Advanced High-Temperature, High-Pressure Transport Reactor Gasification

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Abstract

The U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL) Office of Power Systems Product Management has as its mission to foster the development and deployment of advanced, clean, and affordable fossil-based (coal) power systems. These advanced power systems include the development and demonstration of gasification-based advanced power systems which are an integral part of the Vision 21 program being developed by DOE for the coproduction of power and chemicals. DOE has also been developing advanced gasification systems that lower the capital and operating costs of producing syngas for electricity and chemicals production. A transport reactor has shown potential to be a low-cost syngas producer compared to other gasification systems since its high throughput reduces capital costs. This work directly supports the Power Systems Development Facility (PSDF) utilizing the Kellogg, Brown and Root (KBR) transport reactor located at the Southern Company Services (SCS) Wilsonville, Alabama, site.

Over 2000 hours of operation on nine different coals ranging from bituminous to lignite along with a petroleum coke has been completed to date in the pilot-scale transport reactor development unit (TRDU) at the Energy & Environmental Research Center (EERC). The EERC has established an extensive database on the operation of these various fuels in both air-blown and oxygen-blown modes utilizing a pilot-scale transport reactor gasifier. This database has been useful in determining the effectiveness of design changes on a transport reactor gasifier.

The TRDU has recently been modified by increasing the transport reactor mixing zone length and replacing the J-leg loop seal with an L-valve loop seal. These modifications were undertaken to increase solids circulation rates and solids density in the mixing zone to provide adequate carbon in the bottom of the mixing zone while in oxygen-blown mode. These modifications also increase solid backmixing in the mixing zone, thereby increasing solids residence time and gasifier performance. The effects of different fuel types on both gasifier performance and the operation of the hot-gas filter system have been determined. It has been demonstrated that corrected fuel gas heating values ranging from 105 to 130 Btu/scf have been achieved in air-blown mode while heating values up to 230 Btu/scf on a dry basis have been achieved in oxygen-blown mode. Carbon conversions up to 95% have also been obtained and are highly

dependent on the oxygen/coal ratio. Factors that affect TRDU product gas quality appear to be circulation rate, coal type, temperature, and air/coal and steam/coal ratios. Operating results from the enriched-air and oxygen-blown operation will be utilized to optimize future tests at the PSDF and to make commercial projections about the performance of an oxygen-blown transport reactor in a Vision 21 plant.

Introduction

The objective of the advanced high-temperature, high-pressure transport reactor gasification program at the Energy & Environmental Research Center (EERC) is to demonstrate the successful operation of a transport reactor on a wide variety of fuels.

The goal of the advanced high-temperature, high-pressure transport gasification program at the EERC is to demonstrate acceptable hydrodynamic and gasification performance of the transport reactor development unit (TRDU) under a variety of operating conditions and using a wide range of fuels. The current objectives are focused on understanding and improving the operation of the transport reactor gasifier itself under both air-blown and oxygen-blown conditions. A secondary objective of the program is to demonstrate acceptable performance of hot-gas filter elements on the hot dust-laden fuel gas stream coming from the pilot-scale TRDU system prior to long-term demonstration tests.

Project Description

The pilot-scale TRDU has an exit gas temperature of up to 980°C (1800°F), a gas flow rate of 325 scfm (0.153m³/s), and an operating pressure of 120 psig (9.3 bar). The TRDU system can be divided into three sections: the coal feed section, the TRDU, and the product recovery section. The TRDU proper, as shown in Figure 1, consists of a riser reactor with an expanded mixing zone at the bottom, a disengager, and a primary cyclone and standpipe. The standpipe is connected to the mixing section of the riser by a L-valve transfer line. All of the components in the system are refractory-lined and designed mechanically for 150 psig (11.4 bar) and an internal temperature of 1090°C (2000°F). Detailed design criteria and a comparison to actual operating conditions on the design coal are given in Table 1.

The premixed coal and limestone feed to the transport reactor can be admitted through three nozzles, which are at varying elevations. Two of these nozzles are located near the top of the mixing zone (gasification), and the remaining one is near the bottom of the mixing zone (combustion). During operation of the TRDU, feed is admitted through only one nozzle at a time.

The coal feed is measured by an rpm-controlled metering auger. Oxidant is fed to the reactor through two pairs of nozzles at varying elevations within the mixing zone. For the combustion mode of operation, additional nozzles are provided in the riser for feeding secondary air. Hot solids from the standpipe are circulated into the mixing zone, where they come into contact with the nitrogen and the steam being injected into the J-leg. This feature enables spent char to contact steam prior to the fresh coal feed. This staged gasification process is expected to enhance process
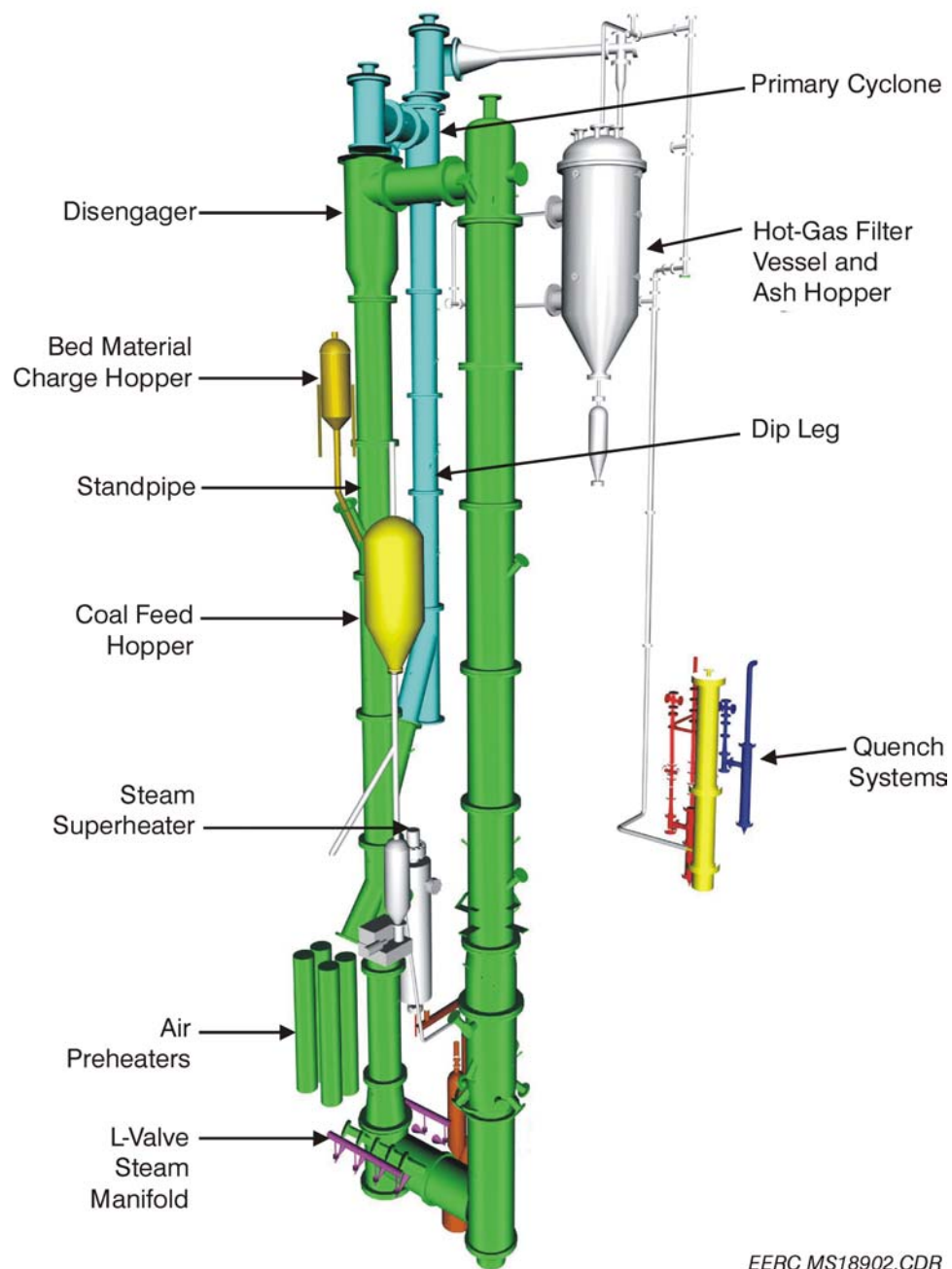
Figure 1. Transport reactor development unit.

Table 1. Summary of TRDU Design and Operation on the Design Coal

Parameter	Design	Actual
Coal	Illinois No. 6	Illinois No. 6
Moisture Content, %	5	8.5
Pressure, psig	120 (9.3 bar)	120 (9.3 bar)
Steam/Coal Ratio	0.34	0.34
Air/Coal Ratio	4.0	2.3
Ca/S Ratio, mole	1.5	2.0
Air Inlet Temperature, °C	427	180
Steam Preheat, °C	537	350
Coal Feed Rate, lb/hr	198 (89.9 kg/hr)	220 (99.9 kg/hr)
Gasifier Temperature, maximum °C	1010	950
ΔT, maximum °C	17	60 to 100
Carbon Conversion,[1] %	>80	76.5
HHV[2] of Fuel Gas, Btu/scf	100	110
Heat Loss as Coal Feed, %	19.5	13
Riser Velocity, ft/sec	31.3	25
Heat Loss, Btu/hr	252,000	320,000
Standpipe Superficial Velocity, ft/sec	0.1	0.38

[1] Carbon Conversion = (wt carbon feed – wt carbon removed)/wt carbon feed * 100.
[2] Higher heating value.

efficiency. Gasification or combustion and desulfurization reactions are carried out in the riser as coal, sorbent, and oxidant (with steam for gasification) flow up the reactor. The solids circulation into the mixing zone is controlled by fluffing gas in the standpipe, J-leg aeration flows, and the solids level in the standpipe.

The riser, disengager, standpipe, and cyclones are equipped with several internal and skin thermocouples. Nitrogen-purged pressure taps are also provided to record differential pressure across the riser, disengager, and the cyclones. The data acquisition and control system scans the data points every one-half second and is saving the process data every 30 s. The bulk of entrained solids leaving the riser is separated from the gas stream in the disengager and circulated back to the riser via the standpipe. A solids stream is withdrawn from the standpipe via an auger to maintain the system's solids inventory. Gas exiting the disengager enters a primary cyclone. Solids from the primary cyclone were collected in a lock hopper for earlier tests. In later tests, the dipleg solids have been recirculated back to the standpipe through the dipleg crossover. Gas exiting this cyclone enters a jacketed-pipe heat exchanger before entering the HGFV. The cleaned gases leaving the HGFV enter a quench system before being depressurized and vented to a flare.

The quench system uses a sieve tower and two direct-contact water scrubbers to act as heat sinks and remove impurities. All entrained solids, any heavy organic vapors, and some water is condensed in the sieve tower, with the first scrubber condensing the remaining water and lighter organics and the second scrubber serving as a backup system. The condensed liquid is separated from the gas stream in a cyclone that also serves as a reservoir. Liquid is pumped either to a

shell-and-tube heat exchanger for reinjection into the sieve tower or scrubbers or down to quench water receiver barrels.

Hot-Gas Filter Vessel

This vessel is designed to handle all of the gas flow from the TRDU at its expected operating conditions. The vessel is approximately 48-in. ID (121.9 cm) and 185 in. (470 cm) long and is designed to handle gas flows of approximately 325 scfm at temperatures up to 815°C (1500°F) and 120 psig (8.3 bar). The refractory has a 28-in. (71.1-cm) ID with a shroud diameter of approximately 22 in. (55.9 cm). The vessel is sized such that it could handle candle filters up to 1.5 m long; however, 1-m candles were utilized in the 540°C (1000°F) gasification tests to date. Candle filters are 2.375-in. (6-cm) OD with a 4-in. (10.2-cm) center line-to-center line spacing. The filter design criteria are summarized in Table 2.

The total number of candles that can be mounted in the current geometry of the HGFV (hot-gas filter vessel) tube sheet is 19. This enables filter face velocities as low as 2.0 ft/min to be tested using 1.5-m candles. Higher face velocities are achieved by using fewer candles. The majority of testing has been performed at a face velocity of approximately 4.0 to 4.5 ft/min. This program has tested a Industrial Filter & Pump (IF&P) ceramic tube sheet and Fibrosic and REECER SiC candles, silicon carbon-coated and SiO_2 ceramic fiber candles from the 3M company, along with sintered metal (iron aluminide) and Vitropore silicon carbon ceramic candles from Pall Advanced Separation Systems Corporation. In addition, granular SiC candles from U.S. Filter/Schumacher and composite candle

Table 2. Design Criteria and Actual Operating Conditions for the Pilot-Scale Hot-Gas Filter Vessel

Operating Conditions	Design	Actual
Inlet Gas Temperature	540°C	450°–580°C
Operating Pressure	150 psig (10.3 bar)	120 psig (8.3 bar)
Volumetric Gas Flow	325 scfm (0.153 m³/s)	350 scfm (0.165 m³/s)
Number of Candles	19 (1 or 1.5 meter)	13 (1 meter)
Candle Spacing	4 in. ₵ to ₵ (10.2 cm)	4 in. ₵ to ₵ (10.2 cm)
Filter Face Velocity	2.5–10 ft/min (1.3 to 2.3 cm/s)	4.5 ft/min (2.3 cm/s)
Particulate Loading	<10,000 ppmw	< 38,000 ppmw
Temperature Drop Across HGFV	<30°C	25°C
Nitrogen Backpulse System Pressure	up to 600 psig (42 bar)	250 to 350 psig (17 to 24 bar)
Backpulse Valve Open Duration	up to 1-s duration	¼-s duration

filters from McDermott Technologies and Honeywell were tested. Later tests have utilized a

metal tube sheet manufactured with expansion cones to allow for thermal stresses. Also, since the metal tubesheet has been installed, candle filter failsafes from Westinghouse Science and Technology Center have also been tested.

The ash letdown system consists of two sets of alternating high-temperature valves with a conical pressure vessel to act as a lock hopper. Additionally, a preheat natural gas burner attached to a lower inlet nozzle on the filter vessel can be used to preheat the filter vessel separately from the TRDU. The hot gas from the burner enters the vessel via a nozzle inlet separate from the dirty gas.

The high-pressure nitrogen backpulse system is capable of backpulsing up to four sets of four or five candle filters with ambient-temperature nitrogen in a time-controlled sequence. The pulse length and volume of nitrogen displaced into the filter vessel is controlled by regulating the pressure (up to 600 psig [42 bar]) of the nitrogen reservoir and controlling the solenoid valve pulse duration. Figure 1 also shows the filter vessel location and process piping in the EERC gasifier tower. Since all of the filter tests are to be completed in the 425°–650°C (800°–1200°F) range, a heat exchanger is used to drop the gas temperature to the desired range. Inserting an existing set of high-temperature valves in the fuel gas heat exchanger can allow for bypassing the filter vessel during TRDU start-up or transients; however, the filter system is rarely bypassed in order to more closely simulate commercial operation. Ports for obtaining hot high-pressure particulate and trace metal samples both upstream and downstream of the filter vessel were added to the filter system piping.

Accomplishments

TRDU Fuel Analysis

The main fuels that have been tested in the TRDU are a Powder River Basin (PRB) subbituminous coal from the Wyodak seam at the Belle Ayr Mine in Gillette, Wyoming; an Illinois No. 6 bituminous coal from Seam 6 of the Baldwin Mine in Baldwin, Illinois; a western bituminous coal mined from the Hiawatha seam at the SUFCo Mine in Salina, Utah; and lignites from the Center, Falkirk, and Freedom Mines in North Dakota. Table 3 shows the proximate, ultimate, higher heating value (HHV), and x-ray fluorescence (XRF) analysis of the Wyodak, Illinois No. 6, SUFCo, and the three lignite coals. All fuels except the lignites were mixed with Plum Run dolomite (PRD) from the Greenfield formation before testing in the TRDU. The lignite gasification tests utilized limestone from the Montana limestone company in Wyoming. Table 4 shows the XRF analyses of the Plum Run dolomite Montana limestones. The calcium-based sorbents were mixed with the respective coals to provide a Ca/S ratio of approximately 2 on a sorbent-only basis for the fuels being gasified (~5 wt% for the PRB and SUFCo coals, 17 wt% for the Illinois No. 6 coal, and 7 to 20 wt% for the North Dakota lignites). Detailed coal analyses show that the Wyodak coal is a high-calcium coal with most of the calcium organically associated and the remaining calcium primarily calcium aluminum phosphate. The mineral sizes are small (70.5 wt% < 10 μm) and are primarily kaolinite and montmorillinite. The Illinois No. 6 coal is a high-iron coal, with most of the iron distributed as pyrite or pyrrhotite with a small

Table 3. Proximate, Ultimate, HHV, and XRF Analyses of Wyodak, Illinois No. 6, SUFCo and Three North Dakota Lignites

	-10-mesh Wyodak Subbituminous Coal	-10-mesh Illinois No. 6 Bituminous Coal	-10-mesh SUFCo Bituminous Coal	-10-mesh Center Lignite Coal	-10-mesh Falkirk Lignite Coal	-10-mesh Freedom Lignite Coal
Proximate Analysis, as run, wt%						
Moisture	20.0	8.5	9.5	35.5	29.50	26.80
Volatile Matter	38.9	36.0	39.1	24.3	30.92	32.52
Fixed Carbon	36.4	44.8	43.8	25.3	27.89	32.48
Ash	4.7	10.7	7.6	14.87	11.69	8.2
Ultimate Analysis, MF[1], wt%						
Carbon	69.06	69.27	77.10	56.72	58.64	62.61
Hydrogen	5.19	5.03	4.61	4.05	4.04	4.25
Nitrogen	0.84	1.1	1.29	0.80	0.81	0.96
Sulfur	0.44	3.55	0.36	1.2	1.06	0.94
Oxygen	18.63	9.34	8.29	19.68	18.87	20.05
Ash	5.85	11.7	8.4	23.1	16.58	11.20
Ash Composition, % as oxides						
Calcium, CaO	26.6	3.2	16.3	8.3	15.5	15.9
Magnesium, MgO	7.0	1.6	3.0	2.8	8.9	5.5
Sodium, Na$_2$O	1.3	1.1	4.6	1.8	0.7	6.0
Silica, SiO$_2$	27.8	53.9	38.3	48.3	41.3	34.6
Aluminum, Al$_2$O$_3$	13.1	21.2	9.3	14.2	12.8	12.6
Ferric, Fe$_2$O$_3$	5.5	13.6	6.1	6.8	4.5	6.6
Titanium, TiO$_2$	1.3	0.9	0.8	0.6	0.5	0.3
Phosphorus, P$_2$O$_5$	1.0	0.2	0.2	0.0	0.2	0.5
Potassium, K$_2$O	0.3	1.9	0.2	2.0	0.4	0.3
Sulfur, SO$_3$	16.0	2.5	21.1	12.2	14.3	17.6
High Heating Value						
MF, Btu/lb	11,700	12,080	12,200	9,446	9,963	10,669
As-Received, Btu/lb	9,750	11,300	11,040	6,093	7,024	7,810

[1] Moisture-free.

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Table 4. XRF Analyses of Plum Run Dolomite and Longview and Montana Limestones

	⁻35-mesh Plum Run Dolomite	⁻35-mesh Montana Limestone
Sorbent Composition, % as oxides		
Calcium, CaO	66.6	73.6
Magnesium, MgO	27.5	0.4
Sodium, Na$_2$O	0.3	0.0
Silica, SiO$_2$	2.7	25.3
Aluminum, Al$_2$O$_3$	1.0	0.0
Ferric, Fe$_2$O$_3$	1.3	0.0
Titanium, TiO$_2$	0.0	0.0
Phosphorus, P$_2$O$_5$	0.0	0.0
Potassium, K$_2$O	0.3	0.3
Sulfur, SO$_3$	0.4	0.4
Loss on Ignition, as run	43.1	36.6

amount of organically associated iron. The high sulfur levels in this coal are the direct result of the high pyrite content. The other primary constituents are aluminum and silica, which are present as quartz or clays. The clays include a substantial amount of potassium aluminosilicate (possibly illite), while the small amount of calcium is present as calcite. The SUFCo coal is a high-calcium coal, with the calcium distributed primarily as calcite along with some organically associated calcium. It has a high quartz and kaolinite content as well. A small amount of iron is found as pyrite, with some of the iron remaining as iron carbonate, submicron iron species, or organically associated iron. SUFCo also has high levels of sodium, which is either associated organically as a salt of a carboxylic group or inorganically as sodium-rich aluminosilicates. The lignite fuels mineral composition is primarily quartz, aluminosilicate clays (especially potassium-based) and pyrite, especially in the case of the Freedom Mine coal. The Kinneman Creek lignite was low in sodium and other alkali species, while the Falkirk lignite was low in sodium but had elevated concentrations of calcium and iron in the ash. The Freedom Mine coal was higher in sodium than the other two lignites and was selected to determine its effect on bed material agglomeration and deposition.

TRDU Operation

A total of 20 test campaigns have been completed to date, with over 2000 hours of operation in gasification on several different fuels. These fuels have ranged from less reactive bituminous coals to the more reactive subbituminous and lignite coals. Operating temperatures have been varied from 815° to 1050°C (1500° to 1900°F), depending on the fuel reactivity and the fuel ashes' propensity to agglomerate. Table 5 summarizes some of the air-blown operating conditions achieved with these various fuels.

Table 5. TRDU Range of Operating Conditions for Air-Blown Operation

Coal Type	Bituminous	Subbituminous	Lignite
Moisture Content, %	8–10	13–24	34–36
Pressure, psig	120	120	120
Steam/Coal Ratio	0.14–0.41	0.23–0.30	0.19–0.29
Air/Coal Ratio	2.8–3.8	2.6–3.5	1.95–2.54
Ca/S Ratio, mole (sorbent only)	2.0	2.0	1.0–2.0
Coal and Sorbent Feed Rate, lb/hr	220–280	220–390	352–492
Avg. Mixing Zone Temp., °C , avg.	920–965	816–900	793–894
HHV of Fuel Gas, Act., Btu/scf	43–75	47–75	46–59
HHV of Fuel Gas, Cor., Btu/scf	70–130	83–126	85–08
Conversion, %	60–87	75–93	70–98
Carbon in Bed, %, Standpipe	5–20	10.5–23.9	7.0–32
Riser Velocity, ft/s	25–34	29.1–40.5	29.1–40.5
Standpipe Velocity, ft/s	0.4–0.45	0.42–0.45	0.39–0.49
Circulation Rate, lb/hr	2235–4200	2960–8200	950–3115
Total Operating Hours	406	980	400

The TRDU has been operated in both air- and oxygen-blown mode on the Illinois No. 6 bituminous coal, the PRB subbituminous coal, and all three North Dakota lignites. Operation on the more reactive western coals has displayed higher carbon conversions and product gas heating values even when operating at lower reactor temperatures than comparable bituminous coal tests. Figure 2 shows the effect of oxygen/coal ratio on carbon conversion and the corrected dry product gas heating value obtained under air-blown conditions for the various fuels. As can be seen, the more reactive lower-rank fuels had higher carbon conversions and corrected dry product gas heating values than the higher-rank bituminous coals. The bituminous coals were operated at higher oxygen/coal ratios than the lower-rank coals since they were operated at higher reactor temperatures in an effort to achieve the same level of steam gasification. For all fuels, carbon conversion increased and corrected dry product gas heating value decreased with increasing oxygen/coal ratio.

Oxygen-blown operation requires the addition of considerable excess steam to maintain the reactor temperatures below the temperature where ash deposition and agglomeration of the circulating ash material becomes a problem. Figure 3 is a plot of both the corrected dry and wet product gas heating values and carbon conversion for the North Dakota lignite fuels under both air- and oxygen-blown conditions. This figure indicates that oxygen-blown operation provides a slightly higher carbon conversion at comparable oxygen/coal ratios. The corrected dry product gas heating value for the oxygen-blown test has a significantly higher heating value than air-blown operation (190 to 225 Btu/scf as compared to 90 to 120 Btu/scf). A comparison of the wet fuel gas heating values shows that air-blown and oxygen-blown gasifiers would have
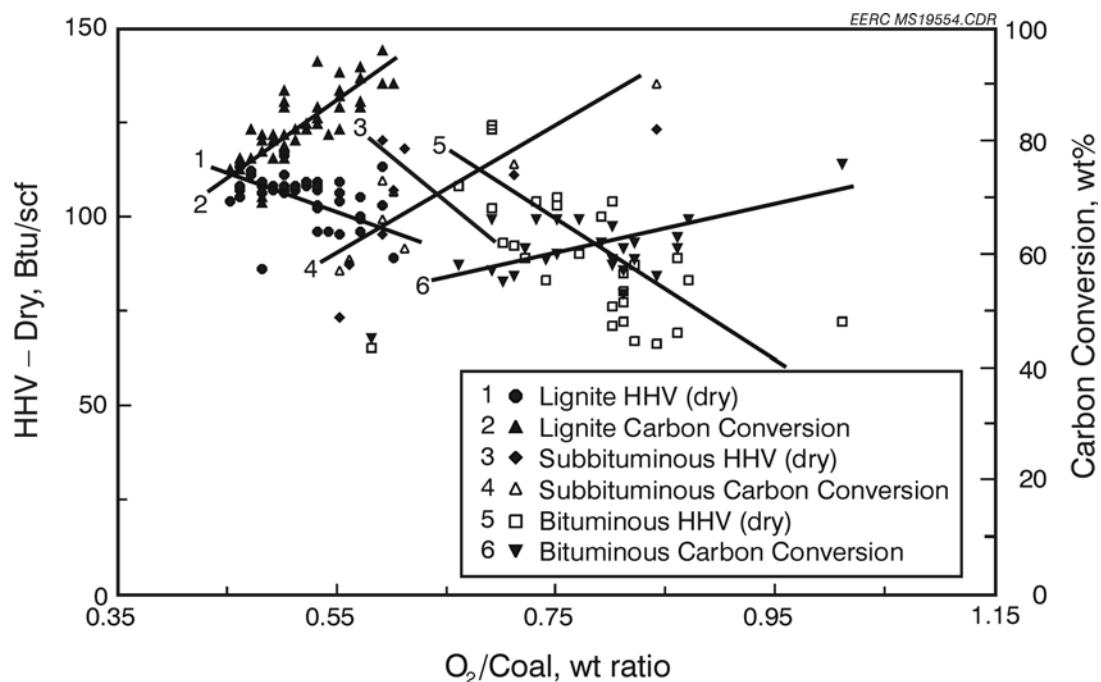
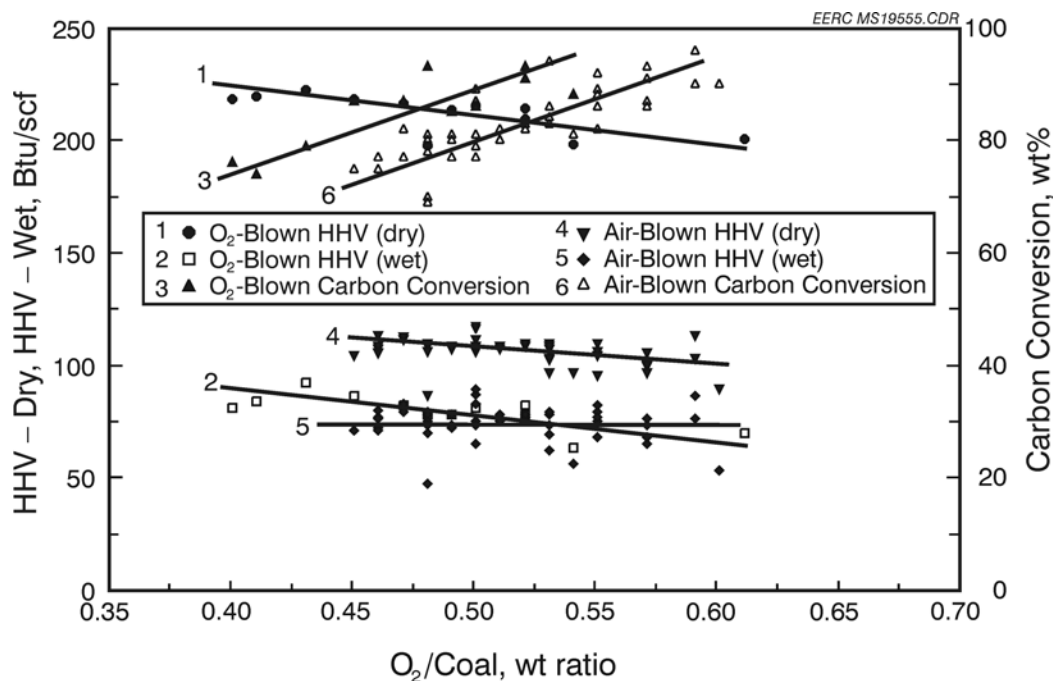Figure 2. TRDU air-blown product gas HHV (dry) and carbon conversion vs. O$_2$/coal.

Figure 3. O$_2$/air-blown lignite HHV and carbon conversion vs. O$_2$/coal ratio.

approximately the same heating value entering the gas turbine combustor because of the high volume of steam addition needed in the oxygen-blown system to prevent circulating bed material from agglomerating and forming deposits on the reactor wall. The similar fuel gas heating values entering a gas turbine make it hard to justify the economics of an oxygen-blown transport reactor strictly for power production. However, for concepts such as a Vision 21 plant in which a gasifier would be operated for both power and fuels/chemicals production, the higher capital and operating costs associated with an oxygen plant may be justified.

Hot-Gas Filter Vessel Operation

Operation of the HGFV during the whole program has mostly tested 1-m-long candles from 3M (SiC-coated fiber), Schumacher Dia-Schumalith 10-20, and Pall Advanced Separations Vitropore 326 and iron aluminide candles along with Westinghouse fail-safes. During the last few tests, 1.5-m-long candles from the 3M Company, Honeywell, and McDermott Technologies and a 1.0-m-long IF&P REECER candle have also been successfully tested. There have been no candle failures in the last 1800 hours of testing. The HGFV has been operated between 410° to 570°C (770° to 1060°F) at a face velocity of approximately 3.8 to 4.5 ft/min. Backpulse operating parameters were 270 to 400 psig reservoir pressure with a ½-s opening time. The average particulate loading going into the HGFV ranged from approximately 4500 up to 38,000 ppm, with a d_{50} between 7 and 22 μm, depending on the fuel type, quantity of sorbent utilized for sulfur control, and whether solids were being recirculated from the dipleg back into the standpipe. Outlet dust loadings have consistently been maintained at 1 ppmw or below. A substantial increase in the "cleaned" filter baseline (from ~25 to 90 in. H_2O) has been observed in some but not all of the tests. This filter ash averaged 40 to 60 wt% carbon and had a low bulk density of approximately 20 lb/ft^3. The small size, the lack of the cohesiveness seen in other filter ashes, and the low density of the ash suggest that a high percentage of the filter cake will be reentrained back onto the filters after they are backpulsed. Off-line cleaning tests were completed which indicated that 20 to 25 in. H_2O of the baseline increase is due to reentrainment of fine filter ash back on the candles and that off-line cleaning times up to 300 s were needed to allow the backpulsed ash to clear the filters. In gasification mode, the pulse frequency has been short, with pulses occurring every 8 to 15 min. This rapid pulsing is thought to be due to the high-carbon, low-density filter cake being able to minimize its porosity on the surface of the candle, thereby resulting in a rapid rise in pressure drop across the filters. Filter permeability tests conducted on cleaned filters that have been removed between tests have shown no significant increase in filter dP over a thousand hours of cumulative testing.

Acknowledgments